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13. ABSTRACT (Maximum 200 words)

An investigation of the nucleation and growth as well as magnetic properties of epitaxial FeAl on AlAs/GaAs(100) is reported. These are the first real space studies of the nucleation, growth and properties of so-called thermodynamically stable films. In-situ RHEED and the first UHV STM measurements were used to characterize the surface. Ex-situ MOKE measurements were used to characterize the magnetic properties. FeAl was found to have an unusual incubation effect over the first 3 bilayers of growth on AlAs. STM images taken at 1 and 3 bilayers examined this effect. After depositing 9 nm and annealing, the films exhibited a 2x2 and/or a 5x5 surface reconstruction. This reconstruction depended upon the anneal temperature and film composition. STM images showed atomic step terraces with step heights roughly corresponding to the height of an FeAl bilayer. Differences were also seen in the surface morphology of the 2-fold and 5-fold surfaces. It was determined that the growth mode was primarily dependent upon growth composition, but had some dependence on the annealed FeAl starting surface. The growth mode changed from monolayer to bilayer closely corresponding to the composition at which FeAl changes from ferromagnetic to non magnetic.

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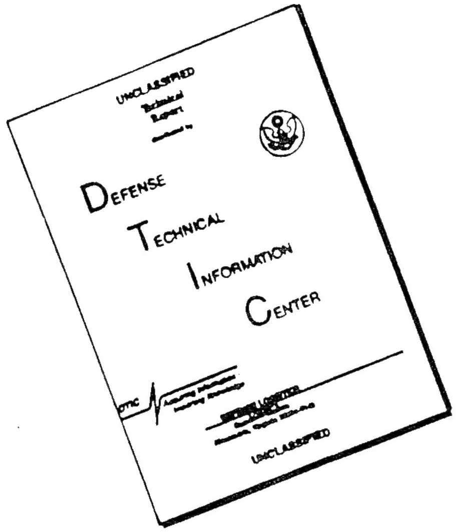

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Contents

1 Abstract

An investigation of the nucleation and growth as well as magnetic properties of epitaxial Fe_xAl_{1-x} on AlAs/GaAs(100) is reported. In-situ RHEED and UHV STM were used to characterize the surface and ex-situ MOKE measurements were used to characterize the magnetic properties. We found that epitaxial films can be grown over a broad composition range $0.5 < x < 0.8$ provided the appropriate nucleation procedure is used, most important of which is the deposition of more than 90Å of Fe_xAl_{1-x} before annealing. Fe_xAl_{1-x} undergoes an unusual incubation effect over the first 3 bilayers of growth on AlAs. STM images taken at 1 and 3 bilayers shed some light on why this incubation effect exists. After depositing 90Å and annealing, the films exhibited a (2×2) and/or a (5×5) surface reconstruction. This reconstuction depended upon the anneal temperature and film composition. STM images of a typical annealed film showed atomic step terraces with step heights roughly corresponding to the height of an Fe_xAl_{1-x} bilayer. Differences were also seen in the surface morphology of the 2-fold and 5-fold surfaces. Growth of Fe_xAl_{1-x} on an annealed Fe_xAl_{1-x} surface produced RHEED oscillations which were was found to occur in 2 distinct modes, monolayer and bilayer. It was determined that this growth mode was primarily dependent upon growth composition, but had some dependence on the annealed Fe_xAl_{1-x} starting surface. The composition at which the growth mode changed from monolayer to bilayer closely corresponded to the composition at which Fe_xAl_{1-x} changes from ferromagnetic to nonmagnetic. Magnetic measurements of several samples confirmed samples above $x = 0.7$ to be ferromagnetic with magnetization in-plane. A compositional dependence on coercivity and saturation magnetization was also found.

2 Introduction

Epitaxial growth of intermetallics on III-V's has been the subject of much research [1] partly because of potential applications in the fabrication of novel electronic and electro-optic devices and partly to overcome problems with creating stable contacts on III-V's. Recently, magnetic intermetallics have begun to receive some attention [2, 3] mainly because of potential applications in the fabrication of novel magnetic devices and their integration with III-V's. Whether depositing magnetic or non-magnetic intermetallics, there is a common rationale for using intermetallics instead of pure metals. Intermetalics are expected to be more thermodynamically stable which is key to achieving abrupt juctions and maintaining epitaxial ordering. In addition, there are a large number of intermetallics, some of which are closely lattice matched to III-V's making them prime candidates for epitaxial growth.

We investigate Fe_xAl_{1-x}on AlAs/GaAs as a continuation of prior efforts by Kuznia and Wowchak to understand it's nucleation and growth. They grew Fe_xAl_{1-x}on pseudomorphic AlAs/InP [5] and later on AlAs/GaAs [4]. In both cases they showed strong evidence that $Fe_xAl_{1-x}$was thermodynamically stable on AlAs to 600 °C over the composition range $0.5 < x < 0.73$. They started with pseudomorphic AlAs/InP substrates because of the relatively small lattice mismatch it has with FeAl (CsCl), -0.9% . They reported seeing several interesting effects. First, the initial nucleation exhibited an incubation period lasting 2 to 3 bilayers in which the intensity of the specular diffraction beam drops to near background and then recovers. Second, growth of Fe_xAl_{1-x}on an annealed Fe_xAl_{1-x}surface resulted in either a bilayer or monolayer growth mode as determined by RHEED oscillations where a monolayer is defined as a single atomic layer with a characteristic thickness of about 1.45Å. A bilayer is two atomic layers and has a charactistic thickness of about 2.90 Å. Lastly, they found surprisingly few differences between growth on AlAs/InP and on AlAs/GaAs even though there is a big difference in lattice mismatch, -0.9% versus 2.9% respectively.

In this report, we addresss several main topics; nucleation, annealing, growth and magnetic properties of Fe_xAl_{1-x}on AlAs/GaAs. In the nucleation section, we report on the use of STM to image the surface during the incubation period. This helps us to provide a more detailed expanation for the incubation effect. In the annealing section, we report on the apparent inability to obtain long range ordering when annealing very thin films and also on the different surface reconstructions that can be abtained after annealing Fe_xAl_{1-x}films of various compositions to temperatures between 550 and 700°C. We also show STM images of these annealed surfaces which confirm previous results of step layer height and terrace size. In the growth section, we report on the factors affecting bilayer and monolayer growth modes and

discuss the possible reasons for two different growth modes. Finally, in the magnetic properties section, we report on MOKE measurements of several films with different compositions in which trends can be observed in the coercivity and saturation magnetization.

3 Experimental

A conventional GaAs MBE system equipped with an Fe e-beam source, specially made to fit in a Kunutzen cell source port, was used for growth. The e-beam source is similar to that described by Jonker et-al [6]. In-situ growth monitoring was performed with a standard RHEED measurement system operating at 10 KeV. Attached to the MBE system was a UHV STM. Samples could be transfered to the UHV STM without exposure to air, allowing for imaging of a clean surface. After growth, MOKE measurements, specifically longitudinal and polar Kerr loops, were performed on several samples in air.

Sample preperation was as follows: Oxides were desorbed from a GaAs (100) N+ Si doped substrate by heating to 620 °C in a Arsenic background of $1 * 10^{-6}$ Torr. A 3000Å GaAs buffer, doped with Si to about $1 \times 10^{18} cm^{-3}$, was then grown. The buffer and substrate were doped to insure that the sample would be conducting for STM measurements. After the GaAs buffer, an AlAs film 10 monolayers thick was deposited. This was intended to act as a diffusion/reaction barrier for the subsequent growth of Fe_xAl_{1-x}. The sample was cooled to 200 °C and removed from the deposition chamber. The Arsenic source was turned down and the remaining Arsenic background was gettered with Ga and Al until the base pressure was below 3×10^{-9} Torr. The Fe and Al sources were then set to give the desired Fe-Al ratio using a quarts crystal deposition rate monitor. The sample was returned to the deposition chamber and heated to 700 °C until an AlAs (3 × 2) pattern appeared. This proceedure was shown to drive off Ga surface contaminates and reduce the Arsenic coverage on the surface [7]. Finally, the sample temperature was brought back down to 200°C and $Fe_xAl_{1-x}$was grown by co-deposition of Fe and Al.

To produce an epitaxial film, we deposited a minimum of 90Å of Fe_xAl_{1-x}on the AlAs surface before annealing to between 550 and 700°C. After annealing, additional $Fe_xAl_{1-x}$was grown at 200 °C by again co-deposititing Fe and Al. After growing roughly 100Å, the film would again be annealed to between 550 and 700°C. This process of growth and annealing could be repeated indefinitely without degridation of the RHEED pattern.

Previous work focused more on the CsCl phase of Fe_xAl_{1-x}which, according to

the bulk Fe-Al phase diagram, exists between $0.5 < x < 0.63$. In our study, we put more emphasis on the BiF_3 phase of Fe_xAl_{1-x} which exist between $0.63 < x < 0.78$. [8] We are interested in this phase because it has a smaller lattice mismatch with GaAs than the CsCl phase, 1.6% versus 2.9addition, Fe_xAl_{1-x} becomes ferromagetic above $x = 0.7$, giving us the ability to study the magnetic properties of these films.

4 Results and Discussion

4.1 Nucleation

As mentioned earlier, the nucleation of Fe_xAl_{1-x} on AlAs exhibits an incubation effect lasting 2-3 bilayers. This effect was found to be more or less independent of growth composition with the exception that the duration of incubation and the rate of recovery was somewhat dependent on composition. In general, higher Fe compositions produced a longer incubation period and slower recovery. This, however, was a small effect and accounted for the incubation lasting up to 5 bilayers instead of 3. Also, after the incubation period, the RHEED intensity weakly oscillated at lower Fe compositions. This did not happen at higher Fe compositions. We did not study these effects in detail. Instead, we attempt to provide some insight into why we get this unusual incubation effect.

Fig. 1 shows a couple of STM images taken at 1 bilayer of growth and 3 bilayers of growth. 1 bilayer of growth corresponds to the point in the incubation where the RHEED pattern intensity reaches a minimum and 3 bilayers correspond to the point where the intensity is almost fully recovered. The first image, Fig 1a, shows a long range scan of the 1 bilayer surface in which we can see large, flat, atomic height terraces. A close range scan, Fig. 1b, shows that the entire surface, including the terraces, are covered with small clusters 40Å in size and 1.8Å high on average. We believe that the clusters are composed of Fe_xAl_{1-x} and the large terraces are due to the underlying AlAs surface. Fig. 1c shows a long range scan of the 3 bilayer surface and Fig. 1d shows a short range scan. It can be seen that the AlAs terraces are no longer visable, but some small features on the surface are still visable. However, they do not appear to be clusters like those shown in Fig 1b. The RHEED patterns at 1 bilayer and 3 bilayer exhibits distinct FeAl 1-fold patterns. At 1 bilayer, the pattern is very dim, but the shape and size of the diffracted beams are nearly identical to the ones at 3 bilayers. The only difference appears to be the intensity.

With the combined information from RHEED and STM we can postulate that the incubation effect is due the the almost immediate formation of amorphous or semi-amorphous FeAl clusters. The amophous or semi-amorphous nature of the

surface would then account for the drop in RHEED intensity. The recovery of the RHEED intensity would then be due to the FeAl ordered phase becoming more energetically favorable than the amorphous phase at around 2 to 3 bilayers.

4.2 Annealing

As we described in the experimental section, we grew at least 90Å of Fe_xAl_{1-x} on AlAs, at low temperatures before annealing. We followed this procedure because atempting to anneal films less than 90Å in thickness commonly resulted in the lack of formation of sharp central peaks in the diffraction pattern. This indicated to us that the film had poor long range order and possibly a rough surface. It was not possible to recover a sharp diffraction pattern by depositing additional $Fe_xAl_{1-x}$and annealing. In addition, it was not possible to obtain RHEED oscillations on such a surface. There was some evidence that the minimum film thickness needed before annealing was dependent upon film composition, however, this was not studied in detail. It is possible that this effect is due to a certain amount of thermodynamic instability of the Fe_xAl_{1-x}films.

We were able to get a good diffraction pattern when annealing films thicker than 90Å. Before annealing, the films exhibited a "fuzzy" (1×1) diffraction pattern. After annealing, the films exhibited either a $(2 \times 2)$and/or a (5×5)diffraction pattern. A sharp (2×2)pattern would appear after annealing to 550 °C for 5 min. The appearance of this pattern was not dependent on composition. However, annealing films to 650-700 °C resulted in the composition dependent formation of a $(2 \times 2)$and/or a (5×5)diffraction pattern. At $x = 0.5$, the film exhibited a pure 2-fold pattern. Between $0.55 < x < 0.7$ the film exhibited a combined 2-fold and 5-fold pattern with the 5-fold pattern becoming more and more dominant at higher Fe concentrations. At or above $x = 0.75$, the film exhibited a pure 5-fold pattern. There was some evidence that the 5-fold pattern is due to excess Fe on the surface. This is discussed in more detail in the growth section.

We looked at the surface of 2 annealed films with STM, one exhibiting a 2-fold diffraction pattern and one exhibiting a 5-fold pattern. Fig 2a shows the image corresponding to the 2-fold pattern. This image reveals a well ordered FeAl surface with average terrace length over 150Å and average step heights of 2.77Å. This step height agrees well the step height of a FeAl bilayer, 2.90Å. Close analysis of the image shows step bunching in many locations where 2 bilayer steps come together to form a single step 5.54Åin height. Fig. 2b shows the image corresponding to the 5-fold pattern. This surface also shows terraces with lengths over 400Å and step heights of 3.14Å on average. Again, the step height is similar in size to that a bilayer step. Comparison of the two images show a dramatic difference in surface

morphology with the 2-fold surface exhibiting rounded corners and the 5-fold surface exhibiting sharp corners.

4.3 Growth

In this section we talk about growth of Fe_xAl_{1-x}on an annealed Fe_xAl_{1-x}surface. As described earlier, we grew at low temperatures, less than 200 °C are got two distinct growth modes as determined by RHEED oscillations, monolayer and bilayer. In general, compositions above $x = 0.7$ resulted in monolayer growth and compositions below $x = 0.7$ resulted in bilayer growth, however the starting surface had some effect on growth mode. By starting surface we mean the annealed Fe_xAl_{1-x}surface upon which we were growing. When attempting to grow on a surface exhibiting a 5-fold diffraction pattern, the first 1 to 15 layers of growth would always be monolayer growth. Assuming the growth composition was below $x = 0.7$, the growth would then transition to bilayer growth after 1 to 15 layers of growth. The thickness at which the growth would transition to bilayer growth was dependent upon composition. At $x = 0.5$ the transition would occur after 1 layer of growth. At $x = 0.6$ the transition would take as much as 10 layers. The closer we got to $x = 0.7$ the longer the transition would take. This effect was in contrast to growth on a 2-fold surface where the transition was quite abrupt. Below $x = 0.7$, the growth would start and remain in bilayer mode. From the results stated above, we concluded that the 5-fold surface was likely Fe rich and that the reason why we would always got monolayer growth on this surface was due to the excess Fe riding the surface and slowly incorporating into the growing layers. If enough Fe was present on the surface, So, growing low Fe compositions on such a surface, the excess Fe would get used up quickly and the growth would transition to bilayer after . At higher Fe compositions, the excess Fe would be used more slowly and the transition to bilayer growth would take longer.

On a 2-fold surface, the transition from monolayer to bilayer occurs at $x = 0.7$ as Kuznia described. However, on a 5-fold surface, the first few layers of growth always occur in a monolayer mode, even at $x = 0.5$ which is well into the bilayer growth region. After several layers of deposition, the growth changes to a bilayer mode. The number of layers needed before a complete transition to bilayer growth is dependent on the growth composition. At $x = 0.5$, the transition from monolayer to bilayer occurs after 1 monolayer of growth. At compositions closer to $x = 0.7$ the transition period may take 40 monolayers or more. The fact the 5-fold surface induces monolayer growth indicates that the surface is Fe rich and the excess Fe incorportates into the growth causing composition to temporarily have more Fe than what is being deposition, thus causing monolayer growth.

4.4 Magnetic Properties

We looked at the magnetic properties of several films to see if they posses magnetic properties that may be of interest for magnetic devices as well as to check if the films posses the expected magnetic properties. Several samples spanning the composition range from ferromagnetic to non-magnetic were measured using MOKE (Magneto-optic Kerr Effect). Figure 4 shows the longitudinal Kerr measurements of these samples. The longitudinal Kerr rotation is a measure of the in-plane magnetization of the film. These measurements show that the transition from magnetic to non-magnetic occurs at a composition around $x = 0.7$, with higher Fe compositions being more magnetic as expected. The magnetic films show 100% in-plane remanence and a coercivity dependence on composition. From polar Kerr measurements, we find the saturation field and maximum Kerr rotation are dependent on composition as well. At $x = 0.2$, $\theta_k = 0.228°$ and $H_s = 16$ KOe whereas for $x = 0.3$, $\theta_k = 0.10°$ and $H_s = 4.5$ KOe. Kerr spectra measurements showed some evidence of structural ordering in the Fe_0.8Al0.2 sample, however the measurements were inconclusive.

5 Conclusion

We report on new findings related to the nucleation, annealing, growth and magnetic properties of epitaxial Fe_xAl_{1-x}on AlAs/GaAs. First, Fe_xAl_{1-x}nucleates on an AlAs (3×2)surface in small oriented crystals. This nucleation behavior may account for the incubation effect observed in the RHEED intensity. Second, it is neccessary to grow a minimum thickness of Fe_xAl_{1-x}on AlAs at low temperatures before annealing in order to obtain a smooth surface. It is unknown, at present, if the reason for the instability of films thinner this minimum thickness is due to a reaction with the underlying AlAs layer or is due to some other process. Third, using the nucleation and growth procedure outlined in the experimental section, highly ordered epitaxial films can be produced over the entire composition range $0.5 < x < 0.8$. The growth mode of Fe_xAl_{1-x}on an annealed Fe_xAl_{1-x}surface is primarily determined by growth composition. Monolayer growth occurs above $x = 0.7$ and bilayer growth occurs below that value. However, the exact growth behavior also depends upon the surface being grown on, where a 5-fold surface can force monolayer growth at least for several layers. Finally, samples with $x > 0.7$ were shown to be ferromagnetic with 100% in-plane magnetization. H_c, M_s and θ_k were found to be dependent on the Fe-Al ratio where all three increase with increasing Fe concentration.

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Figure Captions

Fig.1 RHEED pattern a) and corresponding STM images b) of the initial nucleation after 1 bilayer of deposition on an AlAs (3×2)surface. The STM image shows small structures 40Å in size and 1.8Å high on average.

Fig.2 RHEED pattern showing a sharp 2-fold pattern obtained after annealing to 550 °C and the corresponding STM image showing bilayer high terraces over 150Å in size.

Fig.3 RHEED oscillations showing a) monolayer growth at a composition of $x=0.73$, b) a transition from monolayer to bilayer growth at $x=0.69$ and c) bilayer growth at a composition of $x=0.72$.

Fig.3 Longitudinal Kerr loops of three samples a) $x=0.8$, b) $x=0.7$ and c) $x=0.6$.